



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Patent Application of:

Yasuhiro YOSHIOKA

Group Art Unit: 1752

Application No.: 10/622,668

Examiner: Thorl Chea

Filed: July 21, 2003

For: PHOTOTHERMOGRAPHIC MATERIAL

DECLARATION UNDER 37 C.F.R. §1.132

Honorable Commissioner of Patents and Trademarks

P.O. Box 1450, Alexandria, Virginia 22313-1450

Sir:

I, Yasuhiro YOSHIOKA, do declare and state as follows:

I received a Master's Degree in Chemistry from the Graduate School of The University of Tokyo in March 1980;

I joined Fuji Photo Film Co., Ltd. (now FUJIFILM Corporation) in April 1980, and was engaged in the research of state analysis and reaction analysis of organic materials for silver halide photography from April 1980 to 1989, and in the research of raw materials for silver halide color photographic photosensitive materials and in the research and development of designing of silver

halide color photographic photosensitive materials from 1990 to 1998, and have been engaged in the research of raw materials for silver halide photothermographic photosensitive materials and in the research and development of designing of silver halide photothermographic photosensitive materials from 1999 to present; and

I am familiar with the Office Action of October 9, 2007, and understand that the Examiner has rejected Claims 1, 5, 7, 9, 11, and 13 to 19 under 35 U.S.C. §103(a) as being unpatentable over European Patent No. 1096310 (hereinafter abbreviated as "EP '310") and Oya et al. (U.S. Patent Application No. 2001/0051319 A1).

The following additional comparative experiment was carried out under my supervision in order to make the advantages of the subject matter disclosed and claimed in the above-identified application more clear.

Experiment:

Samples 020' to 026, 030 to 036, 040 to 046, 050 to 056, and 060 to 066 were prepared in the same manner as Sample 008 of Example 1 shown in the specification of the present application, except that the species of the two reducing agents used in Sample 008 and coating amounts

thereof were changed to those shown in the following Tables A' and B. The reducing agents of formula (R1) contained in Samples 020' to 026, 030 to 036, and 040 to 046 were the same (I-6), while the reducing agents of formula (R2) contained therein were varied to I-4, I-9 or I-13. The reducing agents of formula (R2) contained in Samples 050 to 056, and 060 to 066 were the same (I-4), while the reducing agents of formula (R1) contained therein were varied to R1-1 or R1-3 shown in the specification of the present application.

Each of Samples 070, 071, 072, 073, 074, 075, and 076 were prepared in the same manner as Samples 020', 021, 022, 023, 024, 025, or 026 respectively, except that the species of the photosensitive silver halide used in Samples 020' to 026 was changed to that contained in the photosensitive silver halide-containing emulsion "AG-2" as shown in the following Table C.

Each of Samples 080, 081, 082, 083, 084, 085, and 086 were prepared in the same manner as Samples 020', 021, 022, 023, 024, 025, or 026 respectively, except that the species of the photosensitive silver halide used in Samples 020' to 026 was changed to that contained in the photosensitive silver halide-containing emulsion "AG-3" as shown in the following Table C.

Each of Samples 090, 091, 092, 093, 094, 095, and 096

were prepared in the same manner as Samples 020', 021, 022, 023, 024, 025, or 026 respectively, except that the species of the binder used in Samples 020' to 026 was changed to the binder "S-2" as shown in the following Table D.

Each of Samples 100, 101, 102, 103, 104, 105, and 106 were prepared in the same manner as Samples 020', 021, 022, 023, 024, 025, or 026 respectively, except that the species of the binder used in Samples 020' to 026 was changed to the binder "S-3" as shown in the following Table D.

Each of Samples 110, 111, 112, 113, 114, 115, and 116 were prepared in the same manner as Samples 020', 021, 022, 023, 024, 025, or 026 respectively, except that the species of the non-photosensitive organic silver salt (fatty acid silver salt) used in Samples 020' to 026 was changed to that contained in the non-photosensitive organic silver salt dispersion "B-2" as shown in the following Table E.

Each of Samples 120, 121, 122, 123, 124, 125, and 126 were prepared in the same manner as Samples 020', 021, 022, 023, 024, 025, or 026 respectively, except that the species of the non-photosensitive organic silver salt (fatty acid silver salt) used in Samples 020' to 026 was changed to that contained in the non-photosensitive

organic silver salt dispersion "B-3" as shown in the following Table E.

Further, each of Samples 130, 131, 132, 133, 134, 135, and 136 were prepared in the same manner as Samples 020', 021, 022, 023, 024, 025, or 026 respectively, except that the species of the phthalazine compound used in Samples 020' to 026 was changed to the phthalazine compound "F-2" as shown in the following Table F.

The photosensitive silver halide-containing emulsion "AG-1" was the same as the mixed emulsion A for coating solution used in Sample 008 of Example 1 shown in the specification of the present application, and was formed by mixing: 70% by weight of the silver halide emulsion 1 containing a silver bromide iodide particle containing 3.5% by mol iodine uniformly and having an average sphere-equivalent diameter of 0.042 μm ; 15% by weight of the silver halide emulsion 2 containing a pure silver bromide cubic particle having an average sphere-equivalent diameter of 0.080 μm ; and 15% by weight of the silver halide emulsion 3 containing the silver bromide iodide particle containing 3.5% by mol of iodine and having an average sphere-equivalent diameter of 0.034 μm . The photosensitive silver halide-containing emulsion "AG-2" was formed by mixing: 85% by weight of a silver halide emulsion 4 containing a silver bromide iodide particle

containing 4.0% by mol iodine uniformly and having an average sphere-equivalent diameter of 0.036 μm ; and 15% by weight of the silver halide emulsion 2 containing a pure silver bromide cubic particle having an average sphere-equivalent diameter of 0.080 μm . Further, the photosensitive silver halide-containing emulsion "AG-3" was formed by mixing: 85% by weight of a silver halide emulsion 5 containing a silver bromide iodide particle containing 2.5% by mol iodine uniformly and having an average sphere-equivalent diameter of 0.054 μm ; and 15% by weight of the silver halide emulsion 2 containing a pure silver bromide cubic particle having an average sphere-equivalent diameter of 0.080 μm .

In this regard, the silver halide emulsion 4 was prepared in the same manner as the silver halide emulsion 1, except that a solution temperature at particle formation was changed from 30 °C to 28 °C, the amount of 15.3 g of the potassium bromide and the amount of 0.8 g of the potassium iodide in the solution B were changed to 15.2 g and 0.9 g respectively, and the amount of 44.2 g of the potassium bromide and the amount of 2.2 g of the potassium iodide in the solution D were changed to 43.9 g and 2.6 g respectively. Further, the silver halide emulsion 5 was prepared in the same manner as the silver halide emulsion 1, except that a solution temperature at

particle formation was changed from 30 °C to 34 °C, the amount of 15.3 g of the potassium bromide and the amount of 0.8 g of the potassium iodide in the solution B were changed to 15.5 g and 0.6 g respectively, and the amount of 44.2 g of the potassium bromide and the amount of 2.2 g of the potassium iodide in the solution D were changed to 44.6 g and 1.6 g respectively.

The binder "S-1" was the same as the SBR latex used in Sample 008 of Example 1 shown in the specification of the present application, consisting of styrene/butadiene/acrylic acid with molar ratio of 68/29/3, and having an average particle diameter of 90 nm and a glass transition temperature (Tg) of 17°C. The binder "S-2" was a SBR latex consisting of styrene/butadiene/acrylic acid with molar ratio of 71/26/3, and having an average particle diameter of 90 nm and a Tg of 24°C. Further, the binder "S-3" was a SIR latex consisting of styrene/isoprene/acrylic acid with molar ratio of 60/37/3, and having an average particle diameter of 90 nm and a Tg of 14°C.

The non-photosensitive organic silver salt dispersion "B-1" was the same as the fatty acid silver dispersion A used in Example 1 shown in the specification of the present application prepared by using a commercially-available behenic acid EDELOR C22-85R (trade name,

manufactured by Henkel (now renamed as Cognis) and was a mixture of lignoceric acid/behenic acid/arachidic acid/stearic acid with molar ratio of 2/88/8/2) and consists of silver lignocerate/silver behenate/silver arachidate/silver stearate with molar ratio of 2/88/8/2. The non-photosensitive organic silver salt dispersion "B-2" was a fatty acid silver salt dispersion consisting of silver arachidate/silver behenate/silver lignocerate with molar ratio of 2/96/2, and was prepared in the same manner as the non-photosensitive organic silver salt dispersion B-1 except that the EDELOR C22-85R was used after being re-crystallized. Further, the non-photosensitive organic silver salt dispersion "B-3" was a fatty acid silver salt dispersion prepared in the same manner as the non-photosensitive organic silver salt dispersion B-1, except that stearic acid and arachidic acid were added to the EDELOR C22-85R to prepare the fatty acid silver salt contained therein. The fatty acid silver salt contained in the non-photosensitive organic silver salt dispersion B-3 consists of silver stearate/silver arachidate/silver behenate with molar ratio of 8/12/80.

The phthalazine compound "F-1" was the same as the phthalazine compound-1 used in Sample 008 of Example 1 shown in the specification of the present application and is 6-isopropylphthalazine. The phthalazine compound "F-

2" was phthalazine added to Samples 130 to 136 so as to be the same molar amount of the phthalazine compound-1 as that contained in Sample 008 of Example 1.

The thus prepared Samples were subjected to thermal developing treatment and evaluated in terms of tone and tone stability in the same manner as for Example 1 shown in the specification of the present application, except that the a^* and b^* values were measured at points having a concentration of 1.2. The concrete values of a^* , b^* , and the maximum distance R {root of $((\Delta a^*)^2 + (\Delta b^*)^2)$ } are shown in Tables A' to F.

In addition, the plotted a^*b^* ordinates are shown in the attached Figures 1 to 3. In Figures 1 to 3, the plotted a^*b^* ordinates derived from the samples having the same configuration while the contained amount ratio of the reducing agents R1 and R2 being varied are linked with a line. Namely, the a^*b^* ordinates of Samples 020' to 026 (-·-); Samples 030 to 036 (-▲-); Samples 040 to 046 (-■-); Samples 050 to 056 (-◇-); and Samples 060 to 066 (-*-) are respectively linked with a line in Figure 1: the a^*b^* ordinates of Samples 020' to 026 (-·-); Samples 070 to 076 (-▲-); Samples 080 to 086 (-■-); Samples 090 to 096 (-◇-); and Samples 100 to 106 (-*-) are respectively linked with a line in Figure 2: and the a^*b^* ordinates of Samples 020' to 026 (-·-); Samples 110 to 116 (-▲-); Samples 120

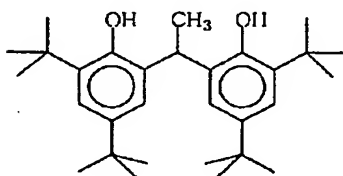
to 126 (-■-); and Samples 130 to 136 (-◇-); are respectively linked with a line in Figure 3.

Ranges of the tone regarded as being preferable or particularly preferable in terms of medical image diagnosis based on the result of a research conducted with image diagnosticians and radiological technicians are also shown in Figures 1 to 3 as the zones enclosed by trapezoids. In the research, each of twenty image diagnosticians and radiological technicians having ten years or more of experience in image diagnosis evaluated twenty test diagnostic images which were prepared by varying compositions to be contained therein and the development conditions such as temperature or time to have the a^* values in the range of 0 to -5 and the b^* values in the range -4 to -12 to determine if each of the test diagnostic images is preferable or not. I declare that it is believed by those skilled in the art at the time that the present invention was invented that the result of the research sufficiently shows the range of tone which is generally recognized as being preferable by persons skilled in the field of image diagnostics.

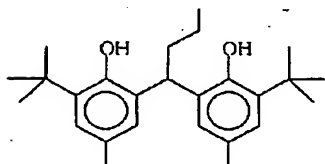
The developing activity of the reducing agent of formula (R2) is judged as being higher than the developing activity of the reducing agent of formula (R1) when the logarithmic value - $\text{Log } E_2$ (E_2 : Exposing amount to give a concentration of 1.5 to a

sample using the reducing agent of formula (R2)) is higher than the logarithmic value $-\text{Log } E_1$ (E_1 : Exposing amount to give a concentration of 1.5 to a sample using the reducing agent of formula (R1)) by 0.02 or larger.

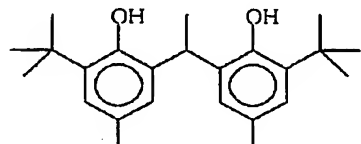
It should be remarked that the reducing agent I-6 shown in EP '310 is within the scope of the reducing agent of formula (R1) of the invention, and the reducing agents I-4, I-9, and I-13 shown in EP '310 are within the scope of the reducing agent of formula (R2) of the invention.



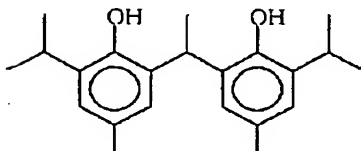
I-6 (corresponding to R1-22 shown in the present application)



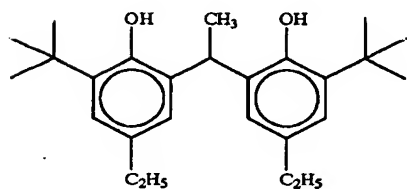
I-4 (corresponding to R2-4 shown in the present application)



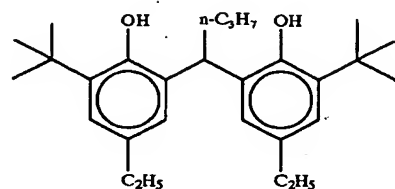
I-9 (corresponding to R2-2 shown in the present application)



I-13



R1-1



R1-3

Table A'

| Sample No. | Reducing agent of formula R1 | | Reducing agent of formula R2 | | Difference in Developing activity of R1 and R2 (R2-R1) | Tone | | | Tone stability | | Remark |
|------------|------------------------------|----------------|------------------------------|----------------|--|-------|-------|-------------------------|--|-------------------------|---------------|
| | Species | Coating amount | Species | Coating amount | | a* | b* | Organoleptic evaluation | Maximum distance ($R = \sqrt{(\Delta a^2 + \Delta b^2)}$) | Organoleptic evaluation | |
| 020' | I-6 | 100% | I-4 | 0% | - | -0.5 | -10.7 | × | 2.18 | × | Comparative |
| 021 | I-6 | 95% | I-4 | 5% | 0.15 | -1.8 | -8.5 | ⊙ | 0.73 | ⊙ | The Invention |
| 022 | I-6 | 90% | I-4 | 10% | 0.15 | -2.15 | -7.5 | ⊙ | 0.46 | ⊙ | The Invention |
| 023 | I-6 | 70% | I-4 | 30% | 0.15 | -2.4 | -6.8 | ○ | 0.78 | ⊙ | The Invention |
| 024 | I-6 | 60% | I-4 | 40% | 0.15 | -2.6 | -6.2 | △ | 1.42 | ○ | The Invention |
| 025 | I-6 | 50% | I-4 | 50% | 0.15 | -2.9 | -5 | × | 2.67 | × | Comparative |
| 026 | I-6 | 40% | I-4 | 60% | 0.15 | -3.1 | -3.5 | × | 3.52 | × | Comparative |
| 030 | I-6 | 100% | I-9 | 0% | - | -0.5 | -10.7 | × | 2.18 | × | Comparative |
| 031 | I-6 | 95% | I-9 | 5% | 0.18 | -2 | -8.8 | ⊙ | 0.64 | ⊙ | The Invention |
| 032 | I-6 | 90% | I-9 | 10% | 0.18 | -2.3 | -8.1 | ⊙ | 0.43 | ⊙ | The Invention |
| 033 | I-6 | 70% | I-9 | 30% | 0.18 | -2.7 | -7.2 | ○ | 0.82 | ○ | The Invention |
| 034 | I-6 | 60% | I-9 | 40% | 0.18 | -2.9 | -6.5 | △ | 0.96 | ○ | The Invention |
| 035 | I-6 | 50% | I-9 | 50% | 0.18 | -3.3 | -5.2 | × | 2.15 | × | Comparative |
| 036 | I-6 | 40% | I-9 | 60% | 0.18 | -3.5 | -4 | × | 3.75 | × | Comparative |
| 040 | I-6 | 100% | I-13 | 0% | - | -0.5 | -10.7 | × | 2.18 | × | Comparative |
| 041 | I-6 | 95% | I-13 | 5% | 0.07 | -1.2 | -9.2 | ○ | 1.12 | ○ | The Invention |
| 042 | I-6 | 90% | I-13 | 10% | 0.07 | -1.6 | -8.2 | ⊙ | 0.88 | ⊙ | The Invention |
| 043 | I-6 | 70% | I-13 | 30% | 0.07 | -1.85 | -7.5 | ⊙ | 0.91 | ⊙ | The Invention |
| 044 | I-6 | 60% | I-13 | 40% | 0.07 | -2 | -6.8 | ○ | 1.33 | ○ | The Invention |
| 045 | I-6 | 50% | I-13 | 50% | 0.07 | -2.15 | -6.1 | △ | 2.42 | × | Comparative |
| 046 | I-6 | 40% | I-13 | 60% | 0.07 | -2.3 | -4.9 | × | 2.96 | × | Comparative |

Table B

| Sample No. | Reducing agent of formula R1 | | Reducing agent of formula R2 | | Difference in Developing activity of R1 and R2 (R2-R1) | Tone | | | Tone stability | | Remark |
|------------|------------------------------|----------------|------------------------------|----------------|--|-------|-------|-------------------------|--|--------------------------|---------------|
| | Species | Coating amount | Species | Coating amount | | a* | b* | Organdep tic evaluation | Maximum distance (R = $\sqrt{(\Delta a^2 + \Delta b^2)}$) | Organdoleptic evaluation | |
| 020' | I-6 | 100% | I-4 | 0% | - | -0.5 | -10.7 | × | 2.18 | × | Comparative |
| 021 | I-6 | 95% | I-4 | 5% | 0.15 | -1.8 | -8.5 | ⊙ | 0.73 | ⊙ | The Invention |
| 022 | I-6 | 90% | I-4 | 10% | 0.15 | -2.15 | -7.5 | ⊙ | 0.46 | ⊙ | The Invention |
| 023 | I-6 | 70% | I-4 | 30% | 0.15 | -2.4 | -6.8 | ○ | 0.78 | ⊙ | The Invention |
| 024 | I-6 | 60% | I-4 | 40% | 0.15 | -2.6 | -6.2 | △ | 1.42 | ○ | The Invention |
| 025 | I-6 | 50% | I-4 | 50% | 0.15 | -2.9 | -5 | × | 2.67 | × | Comparative |
| 026 | I-6 | 40% | I-4 | 60% | 0.15 | -3.1 | -3.5 | × | 3.52 | × | Comparative |
| 050 | R1-1 | 100% | I-4 | 0% | - | -0.8 | -10.6 | × | 2.34 | × | Comparative |
| 051 | R1-1 | 95% | I-4 | 5% | 0.13 | -1.6 | -9.7 | △ | 0.88 | ⊙ | The Invention |
| 052 | R1-1 | 90% | I-4 | 10% | 0.13 | -2 | -9.2 | ○ | 0.65 | ⊙ | The Invention |
| 053 | R1-1 | 70% | I-4 | 30% | 0.13 | -2.4 | -8.7 | ⊙ | 0.82 | ⊙ | The Invention |
| 054 | R1-1 | 60% | I-4 | 40% | 0.13 | -2.9 | -7.9 | ○ | 1.36 | ○ | The Invention |
| 055 | R1-1 | 50% | I-4 | 50% | 0.13 | -3.35 | -6.7 | × | 2.03 | × | Comparative |
| 056 | R1-1 | 40% | I-4 | 60% | 0.13 | -3.8 | -5.3 | × | 2.56 | × | Comparative |
| 060 | R1-3 | 100% | I-4 | 0% | - | -0.6 | -10.6 | × | 2.23 | × | Comparative |
| 061 | R1-3 | 95% | I-4 | 5% | 0.11 | -1.65 | -9.8 | △ | 0.83 | ⊙ | The Invention |
| 062 | R1-3 | 90% | I-4 | 10% | 0.11 | -2.15 | -9.35 | ○ | 0.66 | ⊙ | The Invention |
| 063 | R1-3 | 70% | I-4 | 30% | 0.11 | -2.6 | -8.9 | ⊙ | 0.92 | ⊙ | The Invention |
| 064 | R1-3 | 60% | I-4 | 40% | 0.11 | -2.95 | -8.35 | ○ | 1.45 | ○ | The Invention |
| 065 | R1-3 | 50% | I-4 | 50% | 0.11 | -3.4 | -7.3 | × | 2.15 | × | Comparative |
| 066 | R1-3 | 40% | I-4 | 60% | 0.11 | -3.8 | -6.4 | × | 2.79 | × | Comparative |

Table C

| Sample No. | Photosensitive silver halide | Binder | Non-photosensitive organic silver salt | Phthalazine compound | Difference in Developing activity of R1 and R2 (R2-R1) | Tone | | | Tone stability | | Remark |
|------------|------------------------------|--------|--|----------------------|--|-------|-------|-------------------------|--|-------------------------|---------------|
| | | | | | | a* | b* | Organolectic evaluation | Maximum distance (R = $\sqrt{(\Delta a^*{}^2 + \Delta b^*{}^2)}$) | Organolectic evaluation | |
| 020' | AG-1 | S-1 | B-1 | F-1 | - | -0.5 | -10.7 | × | 2.18 | × | Comparative |
| 021 | AG-1 | S-1 | B-1 | F-1 | 0.15 | -1.8 | -8.5 | ⊙ | 0.73 | ⊙ | The Invention |
| 022 | AG-1 | S-1 | B-1 | F-1 | 0.15 | -2.15 | -7.5 | ⊙ | 0.46 | ⊙ | The Invention |
| 023 | AG-1 | S-1 | B-1 | F-1 | 0.15 | -2.4 | -6.8 | ○ | 0.78 | ⊙ | The Invention |
| 024 | AG-1 | S-1 | B-1 | F-1 | 0.15 | -2.6 | -6.2 | △ | 1.42 | ○ | The Invention |
| 025 | AG-1 | S-1 | B-1 | F-1 | 0.15 | -2.9 | -5 | × | 2.67 | × | Comparative |
| 026 | AG-1 | S-1 | B-1 | F-1 | 0.15 | -3.1 | -3.5 | × | 3.52 | × | Comparative |
| 070 | AG-2 | S-1 | B-1 | F-1 | - | -0.2 | -11.1 | × | 2.38 | × | Comparative |
| 071 | AG-2 | S-1 | B-1 | F-1 | 0.15 | -1.3 | -8.7 | ○ | 0.88 | ⊙ | The Invention |
| 072 | AG-2 | S-1 | B-1 | F-1 | 0.15 | -1.7 | -7.7 | ⊙ | 0.76 | ⊙ | The Invention |
| 073 | AG-2 | S-1 | B-1 | F-1 | 0.15 | -1.9 | -7 | ⊙ | 0.92 | ⊙ | The Invention |
| 074 | AG-2 | S-1 | B-1 | F-1 | 0.15 | -2.05 | -6.2 | △ | 1.41 | ○ | The Invention |
| 075 | AG-2 | S-1 | B-1 | F-1 | 0.15 | -2.3 | -5 | × | 2.75 | × | Comparative |
| 076 | AG-2 | S-1 | B-1 | F-1 | 0.15 | -2.45 | -3.4 | × | 3.63 | × | Comparative |
| 080 | AG-3 | S-1 | B-1 | F-1 | - | -1.1 | -10.5 | × | 2.63 | × | Comparative |
| 081 | AG-3 | S-1 | B-1 | F-1 | 0.15 | -2.2 | -8.6 | ⊙ | 1.22 | ○ | The Invention |
| 082 | AG-3 | S-1 | B-1 | F-1 | 0.15 | -2.45 | -8.1 | ⊙ | 0.86 | ⊙ | The Invention |
| 083 | AG-3 | S-1 | B-1 | F-1 | 0.15 | -2.75 | -7.3 | ○ | 0.88 | ⊙ | The Invention |
| 084 | AG-3 | S-1 | B-1 | F-1 | 0.15 | -3 | -6.5 | △ | 1.27 | ○ | The Invention |
| 085 | AG-3 | S-1 | B-1 | F-1 | 0.15 | -3.25 | -5.6 | × | 2.33 | × | Comparative |
| 086 | AG-3 | S-1 | B-1 | F-1 | 0.15 | -3.45 | -4.6 | × | 2.89 | × | Comparative |

Table D

| Sample No. | Photosensitive silver halide | Binder | Non-photosensitive organic silver salt | Phthalazine compound | Difference in Developing activity of R1 and R2 (R2-R1) | Tone | | | Tone stability | | Remark |
|------------|------------------------------|--------|--|----------------------|--|--------|-------|-------------------------|--|-------------------------|---------------|
| | | | | | | a* | b* | Organoleptic evaluation | Maximum distance (R = $\sqrt{(\Delta a^*{}^2 + \Delta b^*{}^2)}$) | Organoleptic evaluation | |
| 020' | AG-1 | S-1 | B-1 | F-1 | - | -0.5 | -10.7 | × | 2.18 | × | Comparative |
| 021 | AG-1 | S-1 | B-1 | F-1 | 0.15 | -1.8 | -8.5 | ⊙ | 0.73 | ⊙ | The Invention |
| 022 | AG-1 | S-1 | B-1 | F-1 | 0.15 | -2.15 | -7.5 | ⊙ | 0.46 | ⊙ | The Invention |
| 023 | AG-1 | S-1 | B-1 | F-1 | 0.15 | -2.4 | -6.8 | ○ | 0.78 | ⊙ | The Invention |
| 024 | AG-1 | S-1 | B-1 | F-1 | 0.15 | -2.6 | -6.2 | △ | 1.42 | ○ | The Invention |
| 025 | AG-1 | S-1 | B-1 | F-1 | 0.15 | -2.9 | -5 | × | 2.67 | × | Comparative |
| 026 | AG-1 | S-1 | B-1 | F-1 | 0.15 | -3.1 | -3.5 | × | 3.52 | × | Comparative |
| 090 | AG-1 | S-2 | B-1 | F-1 | - | -0.6 | -10.8 | × | 1.97 | △ | Comparative |
| 091 | AG-1 | S-2 | B-1 | F-1 | 0.15 | -1.6 | -8.7 | ⊙ | 0.75 | ⊙ | The Invention |
| 092 | AG-1 | S-2 | B-1 | F-1 | 0.15 | -1.9 | -7.7 | ⊙ | 0.58 | ⊙ | The Invention |
| 093 | AG-1 | S-2 | B-1 | F-1 | 0.15 | -2.15 | -7 | ⊙ | 0.88 | ⊙ | The Invention |
| 094 | AG-1 | S-2 | B-1 | F-1 | 0.15 | -2.3 | -6.4 | △ | 1.31 | ○ | The Invention |
| 095 | AG-1 | S-2 | B-1 | F-1 | 0.15 | -2.6 | -5.2 | × | 1.89 | △ | Comparative |
| 096 | AG-1 | S-2 | B-1 | F-1 | 0.15 | -2.75 | -3.7 | × | 2.46 | × | Comparative |
| 100 | AG-1 | S-3 | B-1 | F-1 | - | -1.1 | -10.9 | × | 1.88 | △ | Comparative |
| 101 | AG-1 | S-3 | B-1 | F-1 | 0.15 | -1.9 | -8.7 | ⊙ | 0.68 | ⊙ | The Invention |
| 102 | AG-1 | S-3 | B-1 | F-1 | 0.15 | -2.3 | -7.6 | ⊙ | 0.52 | ⊙ | The Invention |
| 103 | AG-1 | S-3 | B-1 | F-1 | 0.15 | -2.5 | -6.9 | ○ | 0.83 | ⊙ | The Invention |
| 104 | AG-1 | S-3 | B-1 | F-1 | 0.15 | -2.7 | -6.1 | △ | 1.22 | ○ | The Invention |
| 105 | AG-1 | S-3 | B-1 | F-1 | 0.15 | -2.925 | -5.1 | × | 1.64 | △ | Comparative |
| 106 | AG-1 | S-3 | B-1 | F-1 | 0.15 | -3.15 | -3.6 | × | 2.38 | × | Comparative |

Table E

| Sample No. | Photosensitive silver halide | Binder | Non-photosensitive organic silver salt | Phthalazine compound | Difference in Developing activity of R1 and R2 (R2-R1) | Tone | | | Tone stability | | Remark |
|------------|------------------------------|--------|--|----------------------|--|-------|-------|-------------------------|--|-------------------------|---------------|
| | | | | | | a* | b* | Organoleptic evaluation | Maximum distance (R = $\sqrt{(\Delta a^*{}^2 + \Delta b^*{}^2)}$) | Organoleptic evaluation | |
| 020' | AG-1 | S-1 | B-1 | F-1 | - | -0.5 | -10.7 | × | 2.18 | × | Comparative |
| 021 | AG-1 | S-1 | B-1 | F-1 | 0.15 | -1.8 | -8.5 | ⊙ | 0.73 | ⊙ | The Invention |
| 022 | AG-1 | S-1 | B-1 | F-1 | 0.15 | -2.15 | -7.5 | ⊙ | 0.46 | ⊙ | The Invention |
| 023 | AG-1 | S-1 | B-1 | F-1 | 0.15 | -2.4 | -6.8 | ○ | 0.78 | ⊙ | The Invention |
| 024 | AG-1 | S-1 | B-1 | F-1 | 0.15 | -2.6 | -6.2 | △ | 1.42 | ○ | The Invention |
| 025 | AG-1 | S-1 | B-1 | F-1 | 0.15 | -2.9 | -5 | × | 2.67 | × | Comparative |
| 026 | AG-1 | S-1 | B-1 | F-1 | 0.15 | -3.1 | -3.5 | × | 3.52 | × | Comparative |
| 110 | AG-1 | S-1 | B-2 | F-1 | - | -0.8 | -11.4 | × | 2.19 | × | Comparative |
| 111 | AG-1 | S-1 | B-2 | F-1 | 0.15 | -1.8 | -9.3 | ○ | 0.87 | ○ | The Invention |
| 112 | AG-1 | S-1 | B-2 | F-1 | 0.15 | -2.2 | -8.2 | ⊙ | 0.69 | ⊙ | The Invention |
| 113 | AG-1 | S-1 | B-2 | F-1 | 0.15 | -2.45 | -7.5 | ⊙ | 0.85 | ⊙ | The Invention |
| 114 | AG-1 | S-1 | B-2 | F-1 | 0.15 | -2.8 | -6.3 | △ | 1.33 | ○ | The Invention |
| 115 | AG-1 | S-1 | B-2 | F-1 | 0.15 | -3.05 | -5.3 | × | 2.12 | × | Comparative |
| 116 | AG-1 | S-1 | B-2 | F-1 | 0.15 | -3.3 | -4.1 | × | 2.66 | × | Comparative |
| 120 | AG-1 | S-1 | B-3 | F-1 | - | -0.9 | -10.2 | × | 2.67 | × | Comparative |
| 121 | AG-1 | S-1 | B-3 | F-1 | 0.15 | -1.7 | -8.1 | ⊙ | 1.18 | ○ | The Invention |
| 122 | AG-1 | S-1 | B-3 | F-1 | 0.15 | -1.9 | -7.5 | ⊙ | 0.79 | ⊙ | The Invention |
| 123 | AG-1 | S-1 | B-3 | F-1 | 0.15 | -2.1 | -6.8 | ○ | 1.26 | ○ | The Invention |
| 124 | AG-1 | S-1 | B-3 | F-1 | 0.15 | -2.3 | -6 | △ | 1.68 | △ | The Invention |
| 125 | AG-1 | S-1 | B-3 | F-1 | 0.15 | -2.6 | -4.5 | × | 2.2 | × | Comparative |
| 126 | AG-1 | S-1 | B-3 | F-1 | 0.15 | -2.9 | -2.3 | × | 2.84 | × | Comparative |

Table F

| Sample No. | Photosensitive silver halide | Binder | Non-photosensitive organic silver salt | Phthalazine compound | Difference in Developing activity of R1 and R2 (R2-R1) | Tone | | | Tone stability | | Remark |
|------------|------------------------------|--------|--|----------------------|--|-------|-------|-------------------------|---|-------------------------|---------------|
| | | | | | | a* | b* | Organoleptic evaluation | Maximum distance (R= $\sqrt{(\Delta a^*{}^2 + \Delta b^*{}^2)}$) | Organoleptic evaluation | |
| 020' | AG-1 | S-1 | B-1 | F-1 | - | -0.5 | -10.7 | × | 2.18 | × | Comparative |
| 021 | AG-1 | S-1 | B-1 | F-1 | 0.15 | -1.8 | -8.5 | ⊙ | 0.73 | ⊙ | The Invention |
| 022 | AG-1 | S-1 | B-1 | F-1 | 0.15 | -2.15 | -7.5 | ⊙ | 0.46 | ⊙ | The Invention |
| 023 | AG-1 | S-1 | B-1 | F-1 | 0.15 | -2.4 | -6.8 | ○ | 0.78 | ⊙ | The Invention |
| 024 | AG-1 | S-1 | B-1 | F-1 | 0.15 | -2.6 | -6.2 | △ | 1.42 | ○ | The Invention |
| 025 | AG-1 | S-1 | B-1 | F-1 | 0.15 | -2.9 | -5 | × | 2.67 | × | Comparative |
| 026 | AG-1 | S-1 | B-1 | F-1 | 0.15 | -3.1 | -3.5 | × | 3.52 | × | Comparative |
| 130 | AG-1 | S-1 | B-1 | F-2 | - | -1.9 | -10.8 | × | 2.73 | × | Comparative |
| 131 | AG-1 | S-1 | B-1 | F-2 | 0.15 | -2.1 | -9.7 | △ | 1.64 | △ | The Invention |
| 132 | AG-1 | S-1 | B-1 | F-2 | 0.15 | -2.3 | -9.1 | ○ | 1.18 | ○ | The Invention |
| 133 | AG-1 | S-1 | B-1 | F-2 | 0.15 | -2.6 | -8.5 | ⊙ | 1.23 | ○ | The Invention |
| 134 | AG-1 | S-1 | B-1 | F-2 | 0.15 | -3 | -7.6 | ○ | 1.88 | △ | The Invention |
| 135 | AG-1 | S-1 | B-1 | F-2 | 0.15 | -3.4 | -6.8 | × | 2.68 | × | Comparative |
| 136 | AG-1 | S-1 | B-1 | F-2 | 0.15 | -4.1 | -5.9 | × | 3.79 | × | Comparative |

It can be understood from Figures 1 to 3 that the samples of the present invention (namely, samples having the reducing agent of formula (R1) and the reducing agent of formula (R2) in combination and the coating amount of the reducing agent of formula (R2) is 5 mol% to 40 mol% with respect to the total coating amount of the reducing agents) are within the range preferred by image diagnosticians and radiological technicians, while the comparative examples (namely, samples having the reducing agent of formula (R1) and the reducing agent of formula (R2) in combination and the coating amount of the reducing agent of the formula (R2) is 0 mol%, 50 mol% or 60 mol% with respect to the total coating amount of the reducing agents) are outside the preferred range. The results are well consistent with the results of organoleptic evaluations of tone shown in Tables A' to F.

It can be further understood from Tables A' to F that each of the samples of the present invention has a value R of less than 2, while each of the comparative examples has a value R of at least 2. Since a value R of 2 or more indicates unacceptably large degree of tone difference in view of medical image diagnosis as described in the present specification, the values of R of the comparative examples are outside the preferred range. The results are well consistent with the results of organoleptic

evaluations of tone stability shown in Tables A' to F.

The results exhibited by the samples of the present invention are unexpectedly remarkable in terms of tone and tone stability.

As a person skilled in the art to which the present invention pertains, I believe that the results of the present invention indicated above exhibit unexpectedly remarkable results from a technical viewpoint.

I further declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true, and further, that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

DATE: April 4, 2008

Yasuhiro Yoshioka

Yasuhiro YOSHIOKA

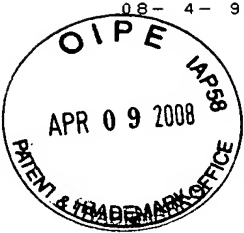


FIG. 1

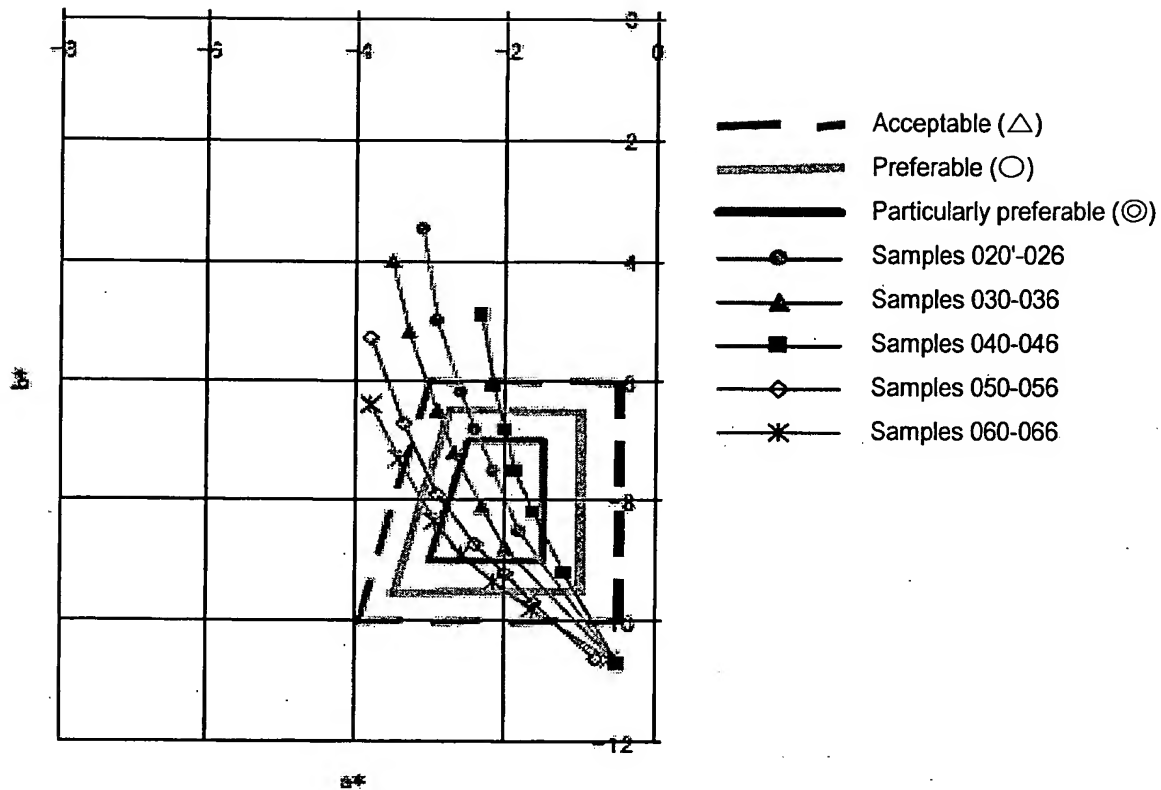


FIG. 2

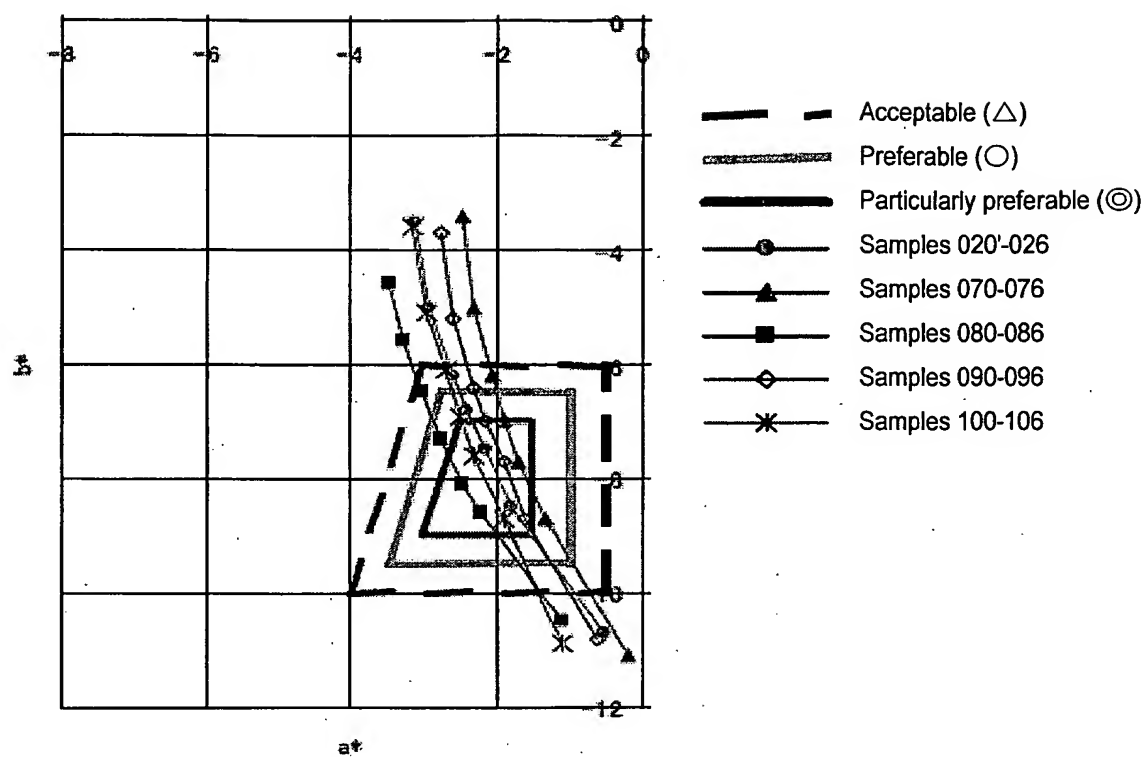


FIG. 3

